THE COMPOSITION OF THE SÃO BRÁS COPPER HOARD IN RELATION TO THE BELL BEAKER METALLURGY IN THE SOUTH-WESTERN IBERIAN PENINSULA*

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A large ceramic vessel was discovered at São Brás (southern Portugal) containing a metallic hoard from the Chalcolithic/Early Bronze Age period. These weapons and tools were characterized by microanalytical techniques as being composed of copper with varying arsenic contents (2.2 ± 1.6 wt%) and minor amounts of lead, bismuth and iron. The collection shows a clear association between daggers and copper with a higher arsenic content, which can be explained by the high status of these silvery alloys. Finally, the compositional distribution of the hoard was compared with the metallurgy of the Bell Beaker and non–Bell Beaker communities inhabiting the south-western Iberian Peninsula.

KEYWORDS: IBERIAN PENINSULA, CHALCOLITHIC, BELL BEAKER, MATERIAL CULTURE, ARSENICAL COPPER, MICRO-EDXRF, MICRO-PIXE

INTRODUCTION

Recent studies concerning the Chalcolithic/Early Bronze Age (*c*.3000–2000 BC) copper metallurgy in the south-western Iberian Peninsula have been depicting a technology with a domestic character, executed in rudimentary metallurgical structures and very dependent on the easily reducible ores available to prehistoric communities (Rovira Llorens 2016). In this sense, this primitive endeavour yielded mostly tools and weapons of simple typology, such as flat axes, saws, chisels, awls or daggers, the composition of which varies from pure copper to high-arsenical copper (Hunt Ortiz 2003; Valério *et al*. 2016a). Moreover, the composition of copper ores and artefacts recovered at archaeological contexts in this region suggests a natural origin for the uneven arsenic amounts of Chalcolithic copper objects (Rovira 2004; Bayona 2008).

During the transition from the second to the third quarter of the third millennium BC, a new cultural identity emerged in Iberian Peninsula—the Bell Beaker Culture—showing distinctive material markers such as novel pottery types and decorations, bone and ivory V-perforated buttons, stone wrist guards, gold ornaments, copper-tanged daggers and Palmela points (Garrido-Pena 2000; García Rivero 2008; García Rivero *et al*. 2016; Guerra Doce and Lettow-Vorbeck 2016). In the south-western region, this culture can be assigned to the chronological interval of 2650/2440−1950/1810 cal BC, 2σ(Soares and Tavares da Silva 2010;

Mataloto *et al.* 2013; Soares 2016). Bell Beaker communities evidence an increased awareness of the aesthetic and useful value of arsenical copper alloys, suggesting the growing importance of metallic items in those prehistoric communities (Valério *et al.* 2016a). However, whether coeval societies shared this metallurgical trend, which consisted of an increased and selective use of arsenical copper alloys, for types that served a certain purpose or an exceptional status is quite another matter.

The São Brás (Serpa) hoard represents an exceptional metal find, being the largest accumulation of Chalcolithic/Early Bronze Age metal in southern Portugal (Soares 2013). The study of such a significant collection constituted a unique opportunity to consolidate our knowledge regarding the early metallurgy of copper. Therefore, the tools and weapons of this hoard were analysed by non-invasive microanalytical techniques to determine the elemental composition without impairing the cultural significance of the collection. The hoard was compared with collections of Chalcolithic (Bell Beaker and non–Bell Beaker) and Middle Bronze Age contexts, allowing a better comprehension of the technological choices of the prehistoric communities inhabiting the south-west of the Iberian Peninsula.

THE SÃO BRÁS HOARD

Artefacts were found in the course of agricultural works carried out during the 1930s and were attributed to the prehistoric settlement of Cerro dos Castelos de São Brás, Serpa (Fig. 1), which was excavated in 1979–80 (Parreira 1983). The São Brás hoard was found inside a complete vessel of globular shape and without any kind of decoration (Soares 2013, 400–7). The metals and ceramic vessel now belong to the collections of the Museum of Archaeology and Ethnography of Setubal District. A close parallel for this type of hoard was found in 1860 at Dolmen de la Pastora, Sevilla (Almagro Basch 1962). The ceramic vase of Dolmen de la Pastora contained 30 javelin arrowheads, which were attributed, following a technological study, to a local production of the Chalcolithic/Early Bronze Age period (Montero-Ruiz and Teneishvili 1996).

The São Brás hoard accounts for a total of about 6.4 kg and 28 artefacts, being mostly composed, as already mentioned, of tools and a few weapons (Soares 2013, 400–7). The absence of ornaments made of copper is common in the early archaeological record of the Iberian Peninsula, a feature that is thought to reflect the lack of aesthetic value of this metal during the Chalcolithic period (Murillo-Barroso and Montero-Ruiz 2012).

The weapons category is represented by two daggers with a midrib and a tanged hilt (CAST. BR.19 and CAST.BR.21) and a flat dagger with a blunt tip (CAST.BR.20) (Fig. 2). It must be emphasized that these tanged daggers are quite different from the typical Bell Beaker types, such as the daggers of Castro dos Ratinhos/Outeiro dos Bravos and Porto Torrão (see Fig. 2). Moreover, a midrib is an uncommon feature for a third millennium BC blade, but Bell Beaker types (Palmela points) with such reinforcement are known in some archaeological contexts in the Iberian Peninsula; that is, at Acebuchal, Sevilla (Hunt Ortiz and Hurtado Pérez 1999), Alcañiz, Teruel (Gabaldón *et al.* 2006) and Coca, Segovia (Gutiérrez Sáez *et al.* 2010). The closest typological and technological parallel for the São Bras hoard is the metallic assemblage of Outeiro de São Bernardo (Moura), also located on the left bank of the Guadiana river and not far from Cerro dos Castelos de São Brás. The site was occupied during the Bell Beaker period (Ciempozuelos-style pottery) and the similarities are very clear with regard to some of the copper types such as the flat dagger and the saws (Cardoso *et al.* 2002).
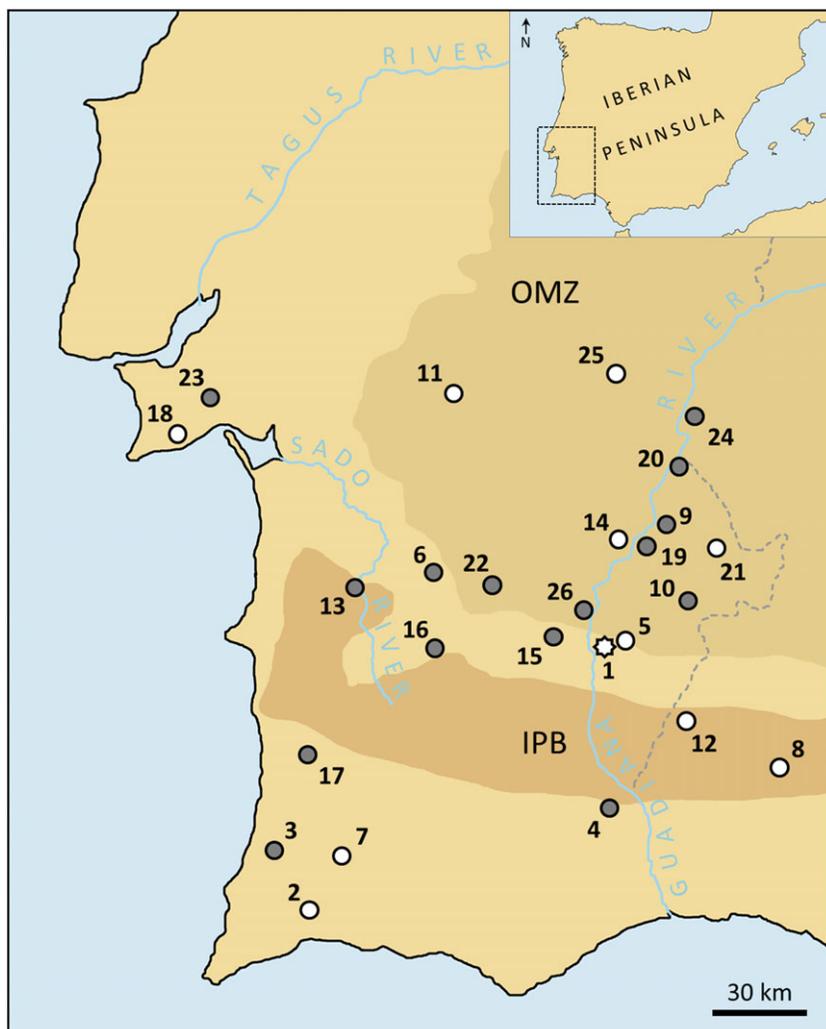
Figure 1   *A map of the south-western Iberian Peninsula with the locations of Chalcolithic sites: 1, São Brás (São Brás 1/ 3 and hoard); 2, Alcalar; 3, Aljezur; 4, Anta do Malhão; 5, Atalaia do Peixoto; 6, Bela Vista 5; 7, Belle France; 8, Cabezo Juré; 9, Castro dos Ratinhos/Outeiro dos Bravos; 10, Corte do Alho; 11, Escoural; 12, La Junta; 13, Lousal; 14, Monte das Aldeias; 15, Monte das Cabeceiras 2/4; 16, Monte do Outeiro; 17, Odemira; 18, Outeiro Redondo; 19, Outeiro de São Bernardo; 20, Porto das Carretas; 21, Porto Mourão; 22, Porto Torrão; 23, Quinta do Anjo; 24, San Blas; 25, São Pedro; 26, Três Moinhos. OMZ, Ossa Morena Zone; IPB, Iberian Pyrite Belt, part of the South Portuguese Zone; the solid circles correspond to archaeological contexts with a significant presence of Bell Beaker cultural markers. [Colour figure can be viewed at wileyonlinelibrary.com]*

The group of tools includes five flat axes and six adzes with rudimentary shapes and curved cutting edges (Fig. 2). Additionally, there is an ingot (CAST.BR.07, Fig. 2) resembling an unfinished axe or adze, but the cutting edge seems to have been thickened by percussion, thus preventing clear identification. These axes and adzes are quite heavy and account for almost 90% of the total weight of the hoard. These simple and heavy items represent a good approach to the eventual long-distance movement of metal and, therefore, early axes are often regarded
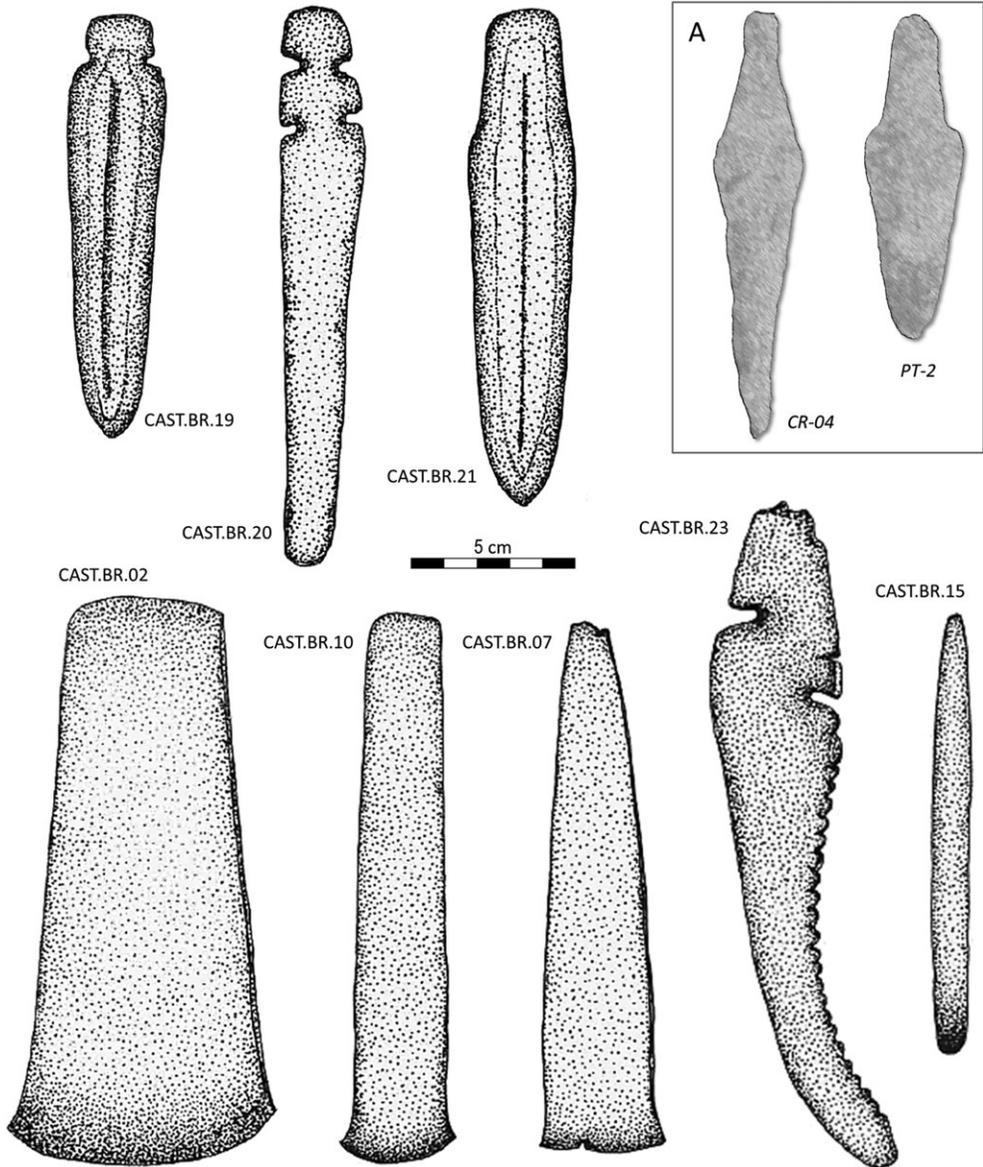
Figure 2   *Artefacts of the São Brás hoard: the daggers (CAST.BR.19, CAST.BR.20 and CAST.BR.21), an axe (CAST. BR.02), an adze (CAST.BR.10), the 'ingot' (CAST.BR.07), a saw (CAST.BR.23) and a chisel (CAST.BR.15) (adapted from Soares 2013). Inset A: two examples of typical Bell Beaker tanged daggers, namely the specimens of Castro dos Ratinhos/ Outeiro dos Bravos (CR-04) and Porto Torrão (PT-2).*

as ingots, not only in the Iberian Peninsula (Müller and Soares 2008; Kunst 2013; Pereira *et al*. 2013a), but also in other regions of Europe (Kienlin *et al*. 2006; Roberts 2009; Delrieu *et al*. 2015). However, it should be stressed that woodworking is the primary function of axes and adzes, and a non-work-hardened replica of an axe has proved to be capable of cutting down a 50-year-old yew tree in a reasonable amount of time (Dolfini 2011).

The remaining tools of the São Brás hoard are six saws and five chisels, typically showing clear use-wear marks such as rounded teeth and asymmetrical bevel edges (there are also two proximal fragments, possibly from saws; Soares 2013, fig. 262, 7 and 8). Some of these saws and two of the daggers described earlier display notches for attachment of the handle. In the south-western Iberian Peninsula, the use of rivets is often attributed to the Middle Bronze Age (Hunt Ortiz 2003). However, the recent discovery of a halberd with rivets in a burial dated to the third quarter of the third millennium BC, at Humanejos (Madrid) (Blasco Bosqued *et al.* 2016), has shown that there is still much to discover regarding third/second millennium BC practices.

METHODOLOGY

This study required a methodology that would not impair the cultural and material significance of the artefacts. Therefore the elemental characterization was made using non-invasive microanalytical techniques, namely energy-dispersive X-ray fluorescence (micro-EDXRF) or particle-induced X-ray emission (micro-PIXE). The latter was only used for analysis of the daggers, as the former technique was unavailable during the short time window in which these specimens could be removed from the exhibition cases at the museum. Nonetheless, it should be mentioned that it had already been established that the analysis of ancient metals by these techniques produces comparable results provided that the analysis is made on a homogeneous sample (Araújo *et al.* 1993; Lyubomirova *et al.* 2015).

Microanalyses of this type of archaeological metal require the removal of the surface corrosion layer over a very small area (3–5 mm diameter) of the specimen. The removal of this surface layer was accomplished by polishing with diamond pastes of increasingly finer grit sizes (15 μm to 1 μm) on the selected area of each artefact. Observations with a Zeiss Discovery V20 stereomicroscope were used to confirm a suitable area for microanalysis, namely a flat surface without the significant presence of corrosion products. After the analytical study, this minute area was protected with benzotriazol (3% m/v in ethanol) and Paraloid B-72 (3% m/v in ethanol).

The micro-EDXRF analyses involved an ArtTAX Pro spectrometer equipped with a 30 W Mo X-ray tube, a focusing polycapillary lens and an electrothermally cooled Si drift detector with a resolution of 160 eV at 5.9 keV (Bronk *et al.* 2001). The prepared area of the artefacts was analysed at three spots (beam spot size of about 70 μm diameter) with a 40 kV tube voltage, a 600 μA current intensity and a livetime of 100 s. The WinAxil software and experimental calibration factors, calculated through the analysis of the British Chemical Standards Phosphor Bronze 551 and BNF Metals Technology Centre Leaded Bronze C50.01, were used for quantitative determinations. Overall, the relative uncertainty of the elemental quantifications is lower than 2% for major elements and lower than 10% for minor elements of interest (Valério *et al.* 2014a). Other elements sometimes also identified in prehistoric copper-based metals are below the detection limit; for example, 0.01 wt% Ni, 0.01 wt% Zn, 0.18 wt% Ag, 0.15 wt% Sn and 0.10 wt% Sb (Vidigal *et al.* 2016).

The micro-PIXE analyses were carried out in a 2.5 MV Van de Graaff accelerator with a 2 MeV proton beam, a beam collimator of 1.5 mm diameter, a beam current intensity of 15 nA for a total accumulated charge of 10 μC and a Si (Li) detector with a resolution of 145 eV at 5.9 keV (Alves *et al.* 2000). Each artefact was analysed at three cleaned spots and the spectral analysis and elemental quantification involved the GUPIXWIN software (Campbell *et al.* 2010). In general, the relative uncertainty of the major elements determination is below 2%, while for trace elements with absolute concentrations around 0.1%, the relative uncertainty is about 5% (Cruz *et al.* 2017).

THE COMPOSITION OF THE HOARD

The elemental analysis of the São Brás hoard has identified copper with varying arsenic contents and occasionally minor amounts of lead and bismuth (Table 1).

The iron content of the São Brás hoard is continuously very low (< 0.05 wt% Fe), similar to other studied artefacts of Chalcolithic contexts in southern Portugal (see a collection with 113 metals in Valério *et al.* 2016a, tables 2–5). In the Iberian Peninsula, prehistoric copper smelting follows the use of rudimentary metallurgical structures, mostly ceramic crucibles, to process the readily available and easily reducible copper minerals, such as carbonates, oxides, phosphates and sulphates (Rovira Llorens 2016). Craddock and Meeks (1987) proposed that the weak reducing atmosphere of prehistoric copper smelting prevented the reduction and incorporation of iron impurities in metallic copper. However, the low iron content of prehistoric copper can also be a consequence of the use of refined copper (Charalambous *et al.* 2014) or the smelting of oxidic copper ores instead of sulphidic copper ores (Klemenc *et al.* 1999; Ashkenazi *et al.* 2012).

As mentioned before, the arsenic content of this collection is quite variable (up to 7.45 wt%), showing an average value of about 2 wt%. The arsenic content among artefacts of the same type is also uneven, but the comparison of average values of different types does highlight some

Table 1 *Elemental composition of copper-based artefacts from São Brás hoard (n.d., not detected)*

	Reference	Weight (g)	Cu (wt%)	As (wt%)	Pb (wt%)	Bi (wt%)	Fe (wt%)
Tools							
Axes	CAST.BR.02	809	99.2	0.73	n.d.	n.d.	< 0.05
	CAST.BR.03	702	97.4	2.54	n.d.	n.d.	< 0.05
	CAST.BR.04	891	97.6	2.36	n.d.	n.d.	< 0.05
	CAST.BR.05	982	97.3	2.68	n.d.	n.d.	< 0.05
	CAST.BR.06	746	98.0	1.95	n.d.	n.d.	< 0.05
Adzes	CAST.BR.08	214	97.8	2.14	n.d.	n.d.	< 0.05
	CAST.BR.09	254	96.8	3.14	n.d.	n.d.	< 0.05
	CAST.BR.10	171	96.7	3.29	n.d.	n.d.	< 0.05
	CAST.BR.11	175	97.8	2.13	n.d.	n.d.	< 0.05
	CAST.BR.12	147	98.8	1.02	0.14	< 0.10	< 0.05
	CAST.BR.13	131	99.0	0.81	0.15	n.d.	< 0.05
Chisels	CAST.BR.14	81	96.7	3.26	n.d.	n.d.	< 0.05
	CAST.BR.15	92	98.0	1.94	n.d.	n.d.	< 0.05
	CAST.BR.16	37	99.2	0.80	n.d.	n.d.	< 0.05
	CAST.BR.17	9	99.8	0.17	n.d.	n.d.	< 0.05
	CAST.BR.18	13	97.8	2.15	n.d.	n.d.	< 0.05
Saws	CAST.BR.22	72	99.9	n.d.	n.d.	n.d.	< 0.05
	CAST.BR.23	55	99.6	0.22	0.16	n.d.	< 0.05
	CAST.BR.24	40	96.1	3.86	n.d.	n.d.	< 0.05
	CAST.BR.25	158	99.1	0.89	n.d.	n.d.	< 0.05
	CAST.BR.26	17	98.7	1.09	n.d.	n.d.	< 0.05
	CAST.BR.27	15	98.7	1.08	n.d.	n.d.	< 0.05
Weapons							
Daggers	CAST.BR.19	32	95.9	4.02	n.d.	n.d.	< 0.05
	CAST.BR.20	19	92.3	7.45	n.d.	0.21	< 0.05
	CAST.BR.21	74	96.0	3.92	n.d.	n.d.	< 0.05
Unknown							
Ingot (?)	CAST.BR.07	357	97.4	2.51	n.d.	n.d.	< 0.05

Table 2 *The arsenic content of different types of copper-based artefacts from the São Brás hoard (average ± standard deviation)*

Type	Sample size	As (wt%)
Hoard	26	2.2 ± 1.6
Axes	5	2.1 ± 0.8
Adzes	6	2.1 ± 1.0
Chisels	5	1.7 ± 1.2
Saws	6	1.2 ± 1.4
Daggers	3	5.1 ± 2.0

interesting features (Table 2). Daggers clearly display higher arsenic contents (5.1 ± 2.0 wt% As) than other types, while saws show an opposing trend (1.2 ± 1.4 wt% As), which is better disclosed (0.7 ± 0.5 wt% As) by discounting one item of uncommonly high content (CAST.BR.24, 3.86 wt%). It has been proposed that tools from domestic contexts, such as these saws, usually endured more recycling cycles than prestige items often recovered at burials (Rovira 2004). The premise follows the depletion of arsenic by oxidation and the evaporation of arsenic trioxide fumes during the melting and post-casting operations, as has been shown by a series of modern experiments (McKerrell and Tylecote 1972).

The similar range of compositions of axes and adzes of the São Brás hoard (0.73−2.68 wt% As and 0.81−3.29 wt% As, respectively) inhibits a possible classification of the 'ingot' CAST. BR07 (2.51 wt% As). Moreover, Chalcolithic copper ingots can show quite uneven compositions, as evidenced by the examples recovered at Cabezo Juré (Huelva)—one ingot made of pure copper (AL3 5035) and another made of high-arsenical copper (AL3 9690, 4.29 wt% As) (Bayona 2008). Additionally, the probable ingot of Outeiro de São Bernardo is the only example made of pure copper, while the remaining tools and weapons of this set show copper with varying amounts of arsenic (Cardoso *et al*. 2002).

The two daggers with a tanged hilt have almost the same composition (~4 wt% As), while the flat dagger has the higher arsenic content of the hoard (7.45 wt% As). The higher arsenic content of the daggers can be associated with different technological aspects. On the one hand, it is known that copper with an increased arsenic content in solid solution can be more hardened by cold work (Lechtman 1996). On the other hand, the formation of an arsenic-rich phase (Cu_3As) gives a silvery colour to arsenical copper alloys. The first concept assumes that the examples with an increased arsenic content are strain hardened and thus show an improved hardness. However, this is contradicted by actual tools and weapons from Chalcolithic sites such as São Pedro (Redondo) (Valério *et al*. 2017), La Junta (Huelva) and Cabezo Juré (Bayona 2008) and Vila Nova de São Pedro (Azambuja) (Pereira *et al*. 2013a). The inability of local metallurgists to control other important factors (e.g., deformation intensity, grain size and the presence of different phases and inclusions) may explain why the strain-hardening advantage of arsenical copper does not seem to have been exploited during the Chalcolithic period.

In other regions, where tools and ornaments made of copper and arsenical copper are commonly present, such as at Tepe Yahya (Iran), the selection of arsenical copper for prestige items is evident (Thornton 2002). In the Iberian Peninsula, the scarcity of copper ornaments during the third millennium BC hinders such a comparison, but some types often considered to be prestige goods can fill this gap. For instance, Chalcolithic daggers on the Italian Peninsula show little wear on their cutting edges, thus being considered cultural items for symbolical rituals (Dolfini 2011).

The selection of silvery raw materials for the São Brás daggers seems quite plausible, as has been also suggested in other instances, such as for third millennium BC settlements in southern Portugal (e.g., São Pedro; Vidigal *et al.* 2016) and Portuguese Estremadura (e.g., Leceia, Müller and Cardoso 2008; and also Vila Nova de São Pedro, Pereira *et al*. 2013a). On the contrary, the five daggers of the Late Chalcolithic monument 3 of Alcalar (Portimão) have lower arsenic contents (0.86–2.3 wt% As; Junghans *et al.* 1960, 1968, 1974), evidencing the irregular features of the primitive metallurgy of the third millennium BC.

COPPER METALLURGY IN THE SOUTH-WESTERN IBERIAN PENINSULA

The lead isotopic data of the archaeological samples indicate the mining of distinct copper sources. For instance, the copper nodules of the settlement of São Pedro imply the smelting of ores of the Ossa Morena Zone (Gauss 2016), while the metallurgical materials at San Blas (Badajoz) point to deposits in the Ossa Morena Zone and the South Portuguese Zone (Hunt Ortiz *et al*. 2009). Moreover, the artefacts of Cabezo Juré (Nocete *et al*. 2004) and a crucible from Aljustrel castle (Pérez Macías *et al*. 2013) suggest the smelting of Iberian Pyrite Belt ores. It has also been established that copper at Portuguese Estremadura settlements—for example, Zambujal (Gauss 2016), Vila Nova de São Pedro (Müller and Soares 2008) and Leceia (Müller and Cardoso 2008)—came from the Ossa Morena Zone, suggesting some sort of trade between those Chalcolithic settlements.

The manufacturing features can be delineated by the microstructural data of about 170 artefacts from Chalcolithic settlements in the south-western Iberian Peninsula, such as São Pedro (Valério *et al*. 2017), La Junta and Cabezo Juré (Bayona 2008), Outeiro Redondo (Pereira *et al*. 2013b), Moita da Ladra (Pereira *et al*. 2017) and Vila Nova de São Pedro (Pereira *et al*. 2013a). The significant occurrence of Cu_2O inclusions indicates a limited control of casting conditions, while the presence of an As-rich phase ($CuAs_3$) in copper with an arsenic content well below the solubility limit further suggests a fast cooling rate after pouring. Concerning the post-casting manufacture, one should first emphasize the absence of 'as-cast' artefacts. The post-casting work commonly included cycles of hammering and annealing, whereas a final work hardening by cold hammering was somewhat less frequent, and mostly applied to sharpen specific areas or improve the surface finishing. Therefore, artefacts show a wide range of hardness without a meaningful correlation with typology or arsenic content, which suggests that the hardening potential of this alloying element was not yet fully understood.

Regarding the composition of those artefacts, the copper-based metallurgy in the south-western Iberian Peninsula was characterized by a conservative technology of variable arsenic contents for almost two millennia (*c*.3000−1200 BC). Apparently, significant developments took place only during the last few centuries of this period—the Middle Bronze Age (MBA) of *c*.1800−1200 BC—not only with the introduction of bronze alloys, but also with an increased usage of arsenical copper alloys (Valério *et al*. 2014b, Valério *et al*. 2016b).

The histogram of arsenic contents of the São Brás hoard shows a trend comparable to that of the Chalcolithic metallurgy in the south-western Iberian Peninsula (Fig. 3). Moreover, both assemblages have a ratio of about 1:1 of artefacts made of copper and arsenical copper. These similar features support the early chronology attributed to the hoard, all the more so because this common pattern is somewhat different from the MBA outline, evidencing a much higher production of arsenical copper alloys (Fig. 3).

Generically, the ascription of an archaeological context to a Bell Beaker community follows the significant existence of cultural markers such as distinctive pottery, Palmela points, stone wrist
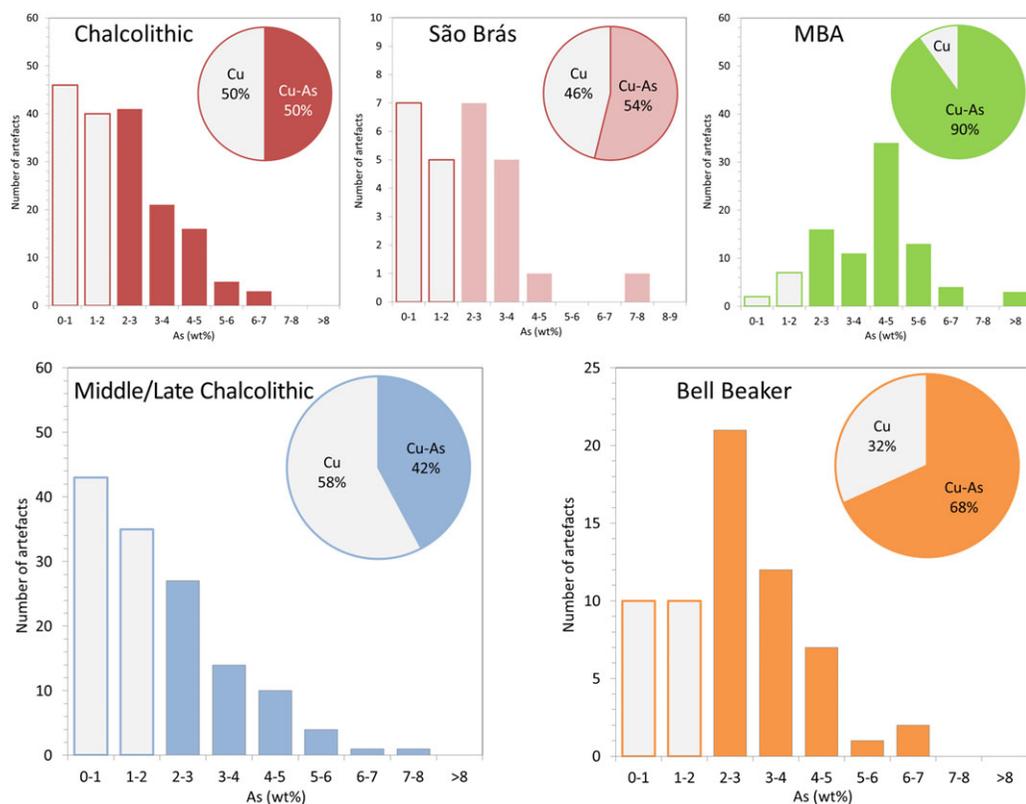
Figure 3   *Histograms and distribution of arsenic contents (Cu: As <2 wt%; Cu-As: As >2 wt%) of copper-based arte-facts from archaeological contexts in the south-western Iberian Penisula. Chalcolithic: Alcalar, Aljezur, Belle France, Quinta do Anjo, Odemira, Lousal and Monte do Outeiro (Junghans et al. 1960, 1968, 1974); Anta do Malhão, Atalaia do Peixoto, Castro dos Ratinhos/Outeiro dos Bravos, Corte do Alho, Monte das Aldeias, Monte das Cabeceiras 2/4, Porto Mourão, Porto Torrão, São Brás 1/3, São Pedro and Três Moinhos (Vidigal et al. 2016; Valério et al. 2016a); Bela Vista 5 (Botainni et al. 2014); Cabezo Juré and La Junta (Bayona 2008); Escoural (Figueiredo et al. 2010); Outeiro Redondo (Pereira et al. 2013b); Porto das Carretas (Valério et al. 2007) and San Blas (Hunt Ortiz et al. 2009). Middle Bronze Age (MBA): Abelheira 1, Horta da Morgadinha, Montinhos 6, Pexem, Torre Velha 12, Vale Frio 2 and Vinha das Caliças 5 (Valério et al. 2016b) and Horta do Folgão, Monte da Cabida 3, Tholos Centirã 2 and Torre Velha 3 (Valério et al. 2014b). For Middle/Late Chalcolithic (non–Bell Beaker) and Bell Beaker and contexts, see Figure 1. [Colour figure can be viewed at wileyonlinelibrary.com]*

guards and V-perforated buttons made of bone or ivory. The São Brás hoard cannot be associated with a Bell Beaker context, since the tanged daggers display an unusual typology and the ceramic vessel does not have the typical Bell Beaker decoration. Following this line of thought, it was pos-sible to compare the composition of artefacts from Bell Beaker contexts (total number of items = 63) and non–Bell Beaker contexts (from now on designated as Middle/Late Chalcolithic contexts: total number of items = 135). The exercise indicates an obvious association of arsenical copper alloys with Bell Beaker contexts in the south-western Iberian Peninsula (Fig. 3). The his-togram of arsenic contents of artefacts from Bell Beaker contexts shows a Gaussian distribution resembling the arsenic content distribution of MBA metals, although with lower levels of arsenic.

The frequency of different types at Bell Beaker and Middle/Late Chalcolithic contexts is somewhat unequal; for example, Bell Beaker contexts have a low occurrence of tools such as

axes, chisels and saws, all of which are common at the remaining Chalcolithic sites. Conse-
quently, the arsenic content of such types must be evaluated to attest to the association of arsen-
ical coppers with Bell Beaker contexts. The assessment made includes a third class comprising
high-arsenical copper alloys (As >4 wt%), which definitely show a silvery colour suitable for
prestige items. Thus, the comparison of the arsenic content distribution of significant types (only
types with more than 10 items were considered) allows some considerations. First, the Bell
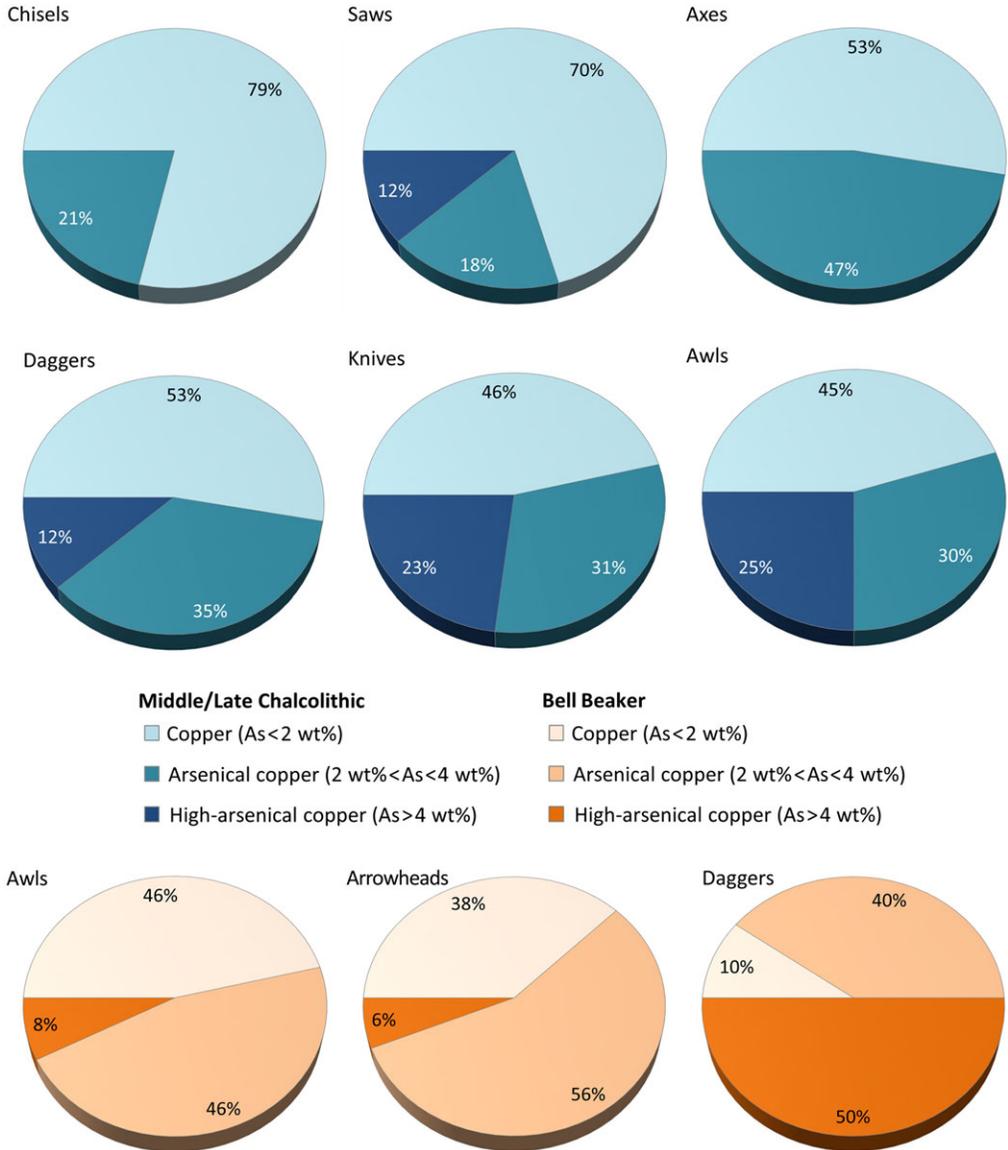

Figure 4   *The distribution of arsenic contents in different types of copper-based artefacts from Middle/Late Chalcolithic
(non–Bell Beaker) and Bell Beaker contexts in the south-western Iberian Penisula (only types with more than 10 items
were considered). [Colour figure can be viewed at wileyonlinelibrary.com]*

Beaker types such as arrowheads and, especially, daggers are predominantly made of arsenical copper or high-arsenical copper (Fig. 4). Moreover, these Bell Beaker daggers show a surprisingly high occurrence of silvery alloys (the high-arsenical copper has a 50% frequency), evidencing a substantial difference to Middle/Late Chalcolithic daggers. Second, most chisels and saws of Middle/Late Chalcolithic contexts are made of copper, a frequency that is reduced to about half of the set of axes, daggers, knives and awls. Actually, these last two types show a higher incidence of high-arsenical copper alloys, suggesting that some specimens could have had a role similar to Bell Beaker daggers among the third millennium BC prestige items.

In sum, for the moment an increased production of arsenical copper alloys is well established among Bell Beaker communities in the south-western Iberian Peninsula, while the distinct composition of daggers suggests that this tendency is independent from the types present in Middle/Late Chalcolithic and Bell Beaker contexts. The compositional discrepancy can hardly be attributed to the copper ores due to the multiple deposits mined along the Ossa Morena Zone and South Portuguese Zone during the Chalcolithic period. However, it should be mentioned that the chronological effect, which can also play a significant role in metallurgical evolution, is very difficult to establish due to the absence of a significant number of artefacts from securely dated archaeological sites.

CONCLUSIONS

The elemental characterization of the São Brás hoard discloses the coexistence of tools and weapons made of copper and arsenical copper, a layout in accordance with third millennium BC metallurgy in the south-western end of the Iberian Peninsula.

The composition of Bell Beaker metal seems to fill the gap between the Chalcolithic and Middle Bronze Age copper-based metallurgies in the region, which show a trend towards increased use of arsenical copper alloys. Some prestige items such as daggers were selectively made of high-arsenical copper alloys, most likely for aesthetic rather than functional purposes. Therefore, the silvery copper would indeed have an aesthetic significance among third millennium BC communities, while the shortage of ornaments can alternatively be explained by a preference for other prestige materials such as gold. The existence of high-arsenical copper daggers in the São Brás hoard implies that this technological enhancement was also present among some of those non–Bell Beaker communities. Overall, the variable composition of the artefacts and the apparent inability to fully exploit the hardening potential of arsenical copper underline the early feature of this copper-based metallurgy that, nevertheless, seems to satisfy consumers' needs throughout the third millennium BC.

Future work should focus on metals from finely dated archaeological contexts to better ascertain the impact of chronology on technological choices of Middle/Late Chalcolithic and Bell Beaker communities. Moreover, additional studies would enable a significant comparison of further artefact types, thus providing a better insight into the early metallurgy in the south-western Iberian Peninsula.

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